15

Total Neutron Counting Instruments and Applicatons

J. Sprinkle

15.1 INTRODUCTION

Total neutron counting instruments are usually unsophisticated instruments that do not measure neutron energy directly. They are simpler than neutron coincidence counters, are capable of detecting smaller quantities of neutron producing materials, and are less sensitive to multiplication effects and efficiency variations. However, they are less capable of determining which isotope or chemical compound produced the neutrons. Sometimes these instruments use partial energy discrimination to reduce the signal from undesired components. More typically, they rely on administrative controls to ensure that only the appropriate materials are assayed.

Many assay instruments for total neutron counting use moderated ^{3}He detectors. The ^{3}He detectors are relatively simple to operate and their reliability is excellent. They can tolerate approximately 10^{13} fast n/cm^2 without serious radiation damage and they provide adequate discrimination against gamma rays in fields less than 1 R/h. Reasonable detection efficiencies can be achieved through careful design. Tubes of 2.5-cm diameter containing 4 atm of ^{3}He have an intrinsic detection efficiency of 90% for thermal neutrons. Detector banks with these tubes placed 5 cm apart can be designed to have absolute detection efficiencies of about 20% for spontaneous fission neutrons. Detector banks have also been built with BF_3 tubes, which provide somewhat better discrimination against gamma rays (see Chapter 13).

This chapter describes several total neutron counting instruments and applications in order of increasing size and complexity. It concludes with examples of three instruments used for special applications: two moisture monitors and an energy-independent long counter.

15.2 THE SHIELDED NEUTRON ASSAY PROBE (SNAP)

The Shielded Neutron Assay Probe (SNAP) is the simplest and most portable of the neutron detectors described in this chapter. An upgraded version, the SNAP-II (Ref. 1), is illustrated in Figure 15.1. SNAP-II consists of two ^{3}He counters (2.5-cm diam, 20-cm active length, 4-atm fill pressure) embedded in a 12.7-cm-diam polyethylene cylinder. The 12.7-cm diameter of the cylinder is optimum for the detection of 1- to 2-MeV fission neutrons. The polyethylene cylinder is wrapped in a thin cadmium sheet to preferentially absorb any background of thermal neutrons. Along its axis is a hole for inserting

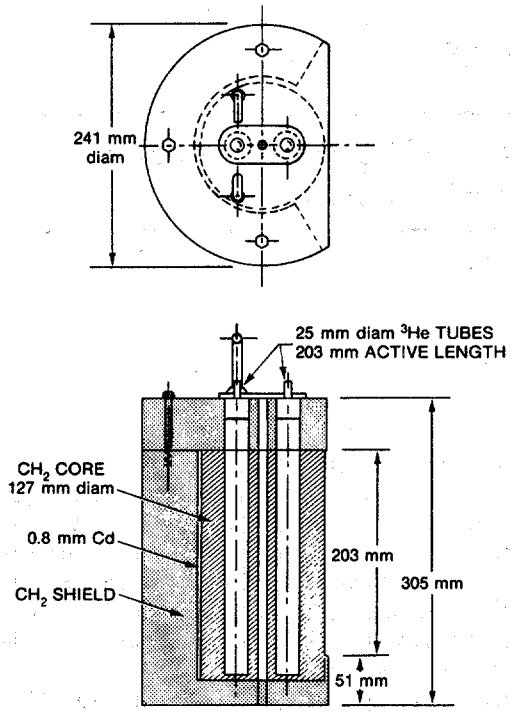

Fig. 15.1 The SNAP-II detector (Ref. 1).

individual reactor fuel rods. This configuration provides an option for high-efficiency assay of plutonium-bearing rods in the hole. Outside the cylinder is a 5.7-cm-thick directional shield whose thickness is limited to ensure portability of the detector. The weight of the SNAP-II is only 10 kg.

With its directional shield, SNAP-II has a viewing angle of 120°. Figure 15.2 shows the relative sensitivity of the detector as a function of angle around the detector axis, as measured with a PuLi source. The front-to-back detection ratio is 4.2 for the PuLi source (average neutron energy of 0.65 MeV) but decreases to 2.2 for a ^{252}Cf spontaneous fission source (average neutron energy of 2.2 MeV).

The intrinsic efficiency of the SNAP-II detector for fission neutrons is approximately 17%, representing a significant improvement over the 10% obtained with the original SNAP detector (Ref. 2). In addition, the active area of the SNAP-II detector is more than twice that of its predecessor; consequently the absolute detection efficiency for a point source at 1 m is improved by a factor of 4.5, to approximately 0.01% (Ref. 1). For other source-to-detector distances, the absolute detection efficiency varies as $(r + a)^{-2}$ where r is measured in centimeters from the detector axis and a is a constant that depends on the diameter of the moderator cylinder. The constant, a, is typically about 3 cm, and r must be greater than 3a for the approximation to be valid. This efficiency function is not valid if the detector is close to surfaces that reflect neutrons (such as a concrete floor). For field exercises that require the use of the SNAP-II in an area surrounded by neutron sources and reflectors, determination of the appropriate background count may be

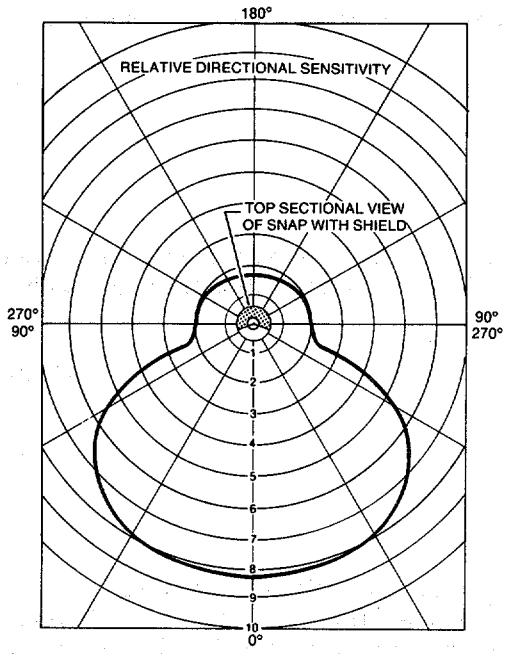

Fig. 15.2 *Directional sensitivity of the SNAP detector, as measured in the midplane with a PuLi source (average energy 0.65 MeV) (Ref. 2).*

difficult. In these instances, it has been helpful to fit a matching wedge of polyethylene into the viewing angle of the detector.

The SNAP-II has been designed for field work and is used in the total neutron counting mode because the signal to be measured is often weak. It is important to use compact, simple electronics. Commercially available packages such as the Eberline SAM I or the Ludlum Model 22 are well suited for this application. These units contain a high-voltage power supply, preamplifier and amplifier boards, discriminators, and scalers in one small portable box.

The SNAP detector provides only limited information about the energy or direction of the neutrons that are detected. It is often used for holdup measurements or verification measurements where good reference standards are not available; examples of such uses follow.

15.2.1 Verification of Plutonium Metal

A SNAP detector has been used to verify the ^{240}Pu content of plutonium metal buttons at the Hanford Works in Richland, Washington (Ref. 3). The major neutron source in metallic plutonium is the spontaneous fission of ^{240}Pu. In addition, buttons containing 2000 g of plutonium exhibit significant multiplication effects. These effects can increase the totals count rate by as much as 90% and are dependent on the button geometry.

At Hanford, the measured SNAP response, T total neutrons/s, was represented as

$$T(n/s) = k\ m\ I\ F \tag{15-1}$$

where k = calibration constant
m = elemental plutonium mass
I = ^{240}Pu mass fraction

and F is a factor that accounts for multiplication within the button. F was determined empirically by fitting a series of Monte Carlo calculations of multiplication within buttons, with the following result:

$$F = 1 + (1 - aI)m/b \tag{15-2}$$

$$a = \frac{619.5\,(d - 2.673)^2 + 1801}{b} \tag{15-3}$$

$$b = 417.8\ (d - 1.333)^2 + 1757 \tag{15-4}$$

where d is the button diameter in inches, and a and b are also given in inches.

Equation 15-1 can be solved for either m or I. At Hanford, one button was chosen as the "standard" and the remaining were assayed as unknowns. A statistical analysis of the assay of 248 buttons yielded 8 outliers. The ^{240}Pu content of the other 240 buttons was verified. The mass range of the buttons was 1453 to 2204 g and the ^{240}Pu mass fraction ranged from 4.6 to 18.1%. The measurement uncertainty was 2% (1σ), and the precision in 30-s counts was better than 1%.

15.2.2 Verification of UF_6 Cylinders

SNAP detectors have been used at enriched uranium production and storage facilities to verify the contents of UF_6 cylinders (Ref. 4). In UF_6, neutrons are produced by spontaneous fission of ^{238}U and by the ^{19}F(α,n)^{22}Na reaction. The dominant alpha-particle emitter is ^{234}U. In natural UF_6, 80% of the neutrons are due to (α,n) reactions and the rest to spontaneous fission. As the enrichment increases, the contribution of the (α,n) reaction increases sharply. In particular, as the enrichment increases from 3 to 90%, the neutron production rate increases by a factor of 30. Consequently the totals neutron count rate is a sensitive measure of the UF_6 mass and enrichment. Neutrons penetrate UF_6 quite well but are subject to absorption and multiplication effects. Table 15-1 lists the calculated neutron leakage fraction from various cylinders. At the higher enrichments, multiplication effects begin to dominate over absorption effects.

To verify the contents of a UF_6 cylinder, the SNAP detector is placed adjacent to the cylinder and parallel to its axis, at a position midway between the cylinder ends. This geometry is less position dependent, less sensitive to the cylinder's fill height, and more convenient for the operator than a geometry that requires the operator to hold the SNAP against the end of the cylinder. Backgrounds caused by neighboring cylinders are often large. Reasonable background estimates (at sea level) are obtained by aiming the

Table 15-1. Neutron leakage fraction from various cylinders

Cylinder Type	UF_6 Mass (kg)	^{235}U Enrichment (wt%)	Leakage Fraction
14 ton	12,700	0.71	0.55
30B	1,500	0.71	0.80
12B	140	3.00	0.97
5A	15	10.00	1.00
5A	15	35.00	1.00
2S	1	65.00	1.01
2S	1	90.00	1.02

detector upward toward the sky rather than downward toward the cylinder. The response from cylinders at the edge of a large cylinder array may be smaller than the response near the center of the array. For cylinders containing UF_6 of different enrichments, the total neutron count rate T is given by

$$T = [cf(234) + df(238)]\ m \tag{15-5}$$

where m is the total uranium mass, c and d are empirical calibration constants, and f(234) and f(238) are the isotopic mass fractions of ^{234}U and ^{238}U, respectively.

For low enrichments, f(238) is nearly constant, and the ratio f(234)/f(235) is very nearly constant. Then Equation 15-5 can be simplified to

$$T = [e + gf(235)]\ m \tag{15-6}$$

where e and g are the calibration constants. Clearly, if the isotopic composition of the UF_6 is the same for all cylinders, Equation 15-6 can be further simplified to $T = hm$, with h being the single calibration constant.

Measurement uncertainties of 5% are typical for 2 1/2-, 10-, and 14-ton cylinders. With 60- to 120-s counting times, statistics do not contribute appreciably to these uncertainties. Experience in the field suggests that each cylinder size requires a different calibration, perhaps because different solid angles are subtended at the detector by the various cylinders. If the cylinder fill heights vary by 50%, the uncertainties will increase to approximately 10%. Measurements made with the SNAP detector can help identify solid residues in emptied cylinders (heels), but the small amount of material involved causes poor precision in the results and may require long count times, on the order of 1000 s.

15.2.3 Holdup Measurements

SNAP detectors have been used in nuclear fuel-cycle facilities to measure the holdup of nuclear material (Refs. 5 through 7). The material to be measured must be a strong neutron source. All forms of plutonium qualify, but uranium must be enriched and in a matrix suitable for (α,n) reactions so that the signal will be large enough to be useful.

The primary advantage of using neutron-based techniques for holdup measurements is the penetrability of the neutrons. They can be detected from pumps, furnaces, and other heavy equipment that is too dense to permit the escape of gamma radiation. The disadvantages of neutron techniques are their lack of spatial resolution, their lack of isotopic specificity, and their sensitivity to matrix effects. The matrix effects include reflection, multiplication, moderation by moisture, and (α,n) reactions in low-Z materials. Most of these effects are present to some degree in all holdup situations and make the interpretation of neutron measurements difficult. Use of a combination of neutron and gamma-ray measurements is usually the most reliable approach.

Calibration of the SNAP detector for holdup measurements requires standards with isotopic and chemical compositions that are similar to those of the material held up in process, for the reasons mentioned above. To calibrate for a particular geometry, it is helpful to use mockups of the actual equipment. As in all process holdup measurements, 25 to 50% uncertainties are typical. Additional information on holdup measurements with neutrons is included in Section 20.6 of Chapter 20.

15.2.4 Other Applications

Simple detectors like the SNAP can be used as neutron monitors in and around storage vaults and reactors (Refs. 8 and 9). The monitors are used to check for a constant neutron flux. They may have hardened electronics, simple mass-produced electronics, or bare ^{3}He tubes, depending on the application.

15.3 SLAB DETECTORS

Slab detectors contain an array of thermal neutron detectors inserted into a slab of moderating material. They are larger and heavier than SNAP detectors but provide higher detection efficiencies and better directionality if heavier shielding and collimators are added. Slab detectors are typically operated with a combination of standard NIM electronics modules (such as high voltage, amplifier, single-channel analyzer, and scaler) and customized preamplifiers. The preamplifiers are small enough to fit inside the junction box that contains the connections to the tubes. Pulse-height analysis with a multichannel analyzer is sometimes used, but scalers and single-channel analyzers are more frequently found in routine operation.

Slab detectors often consist of ^{3}He tubes placed parallel to each other in a moderator block (Ref. 10). The size and number of tubes vary with the application. Figure 15.3 shows a slab detector that contains nine ^{3}He tubes (2.5-cm diam, 4-atm fill pressure) embedded in a 10-cm-thick polyethylene slab. The junction box holds the tubes rigidly and encloses the high-voltage buss wire and preamplifier in an air-tight, electrically shielded space. For some applications the polyethylene slab is covered with a thin cadmium sheet to absorb thermal neutrons.

15.3.1 Monitoring of UF_6 Enrichment

A slab detector can be operated unshielded or it can be placed inside a shield to obtain direction-sensitive response. Figure 15.4 illustrates a shielded, directional slab detector

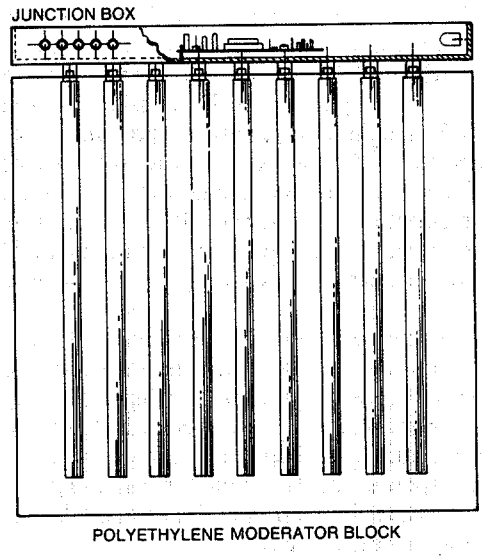

Fig. 15.3 *Side view of slab detector with nine 2.5-cm-diam ^{3}He proportional counters. The junction box contains a preamplifier board.*

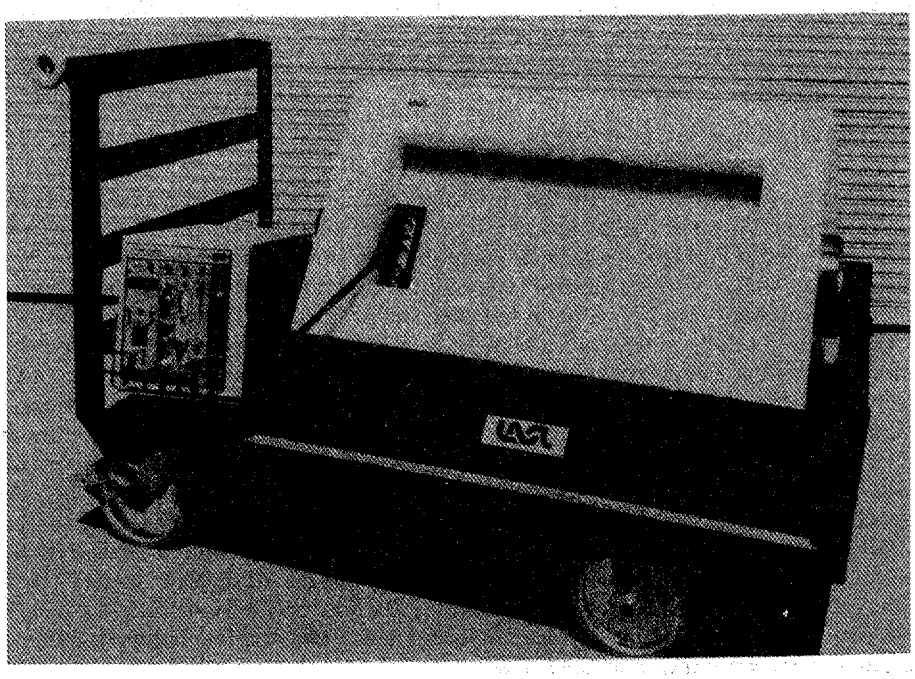

Fig. 15.4 *A slab detector with directional shielding designed for monitoring UF_6 enrichment.*

designed for monitoring UF_6 enrichment. It has a viewing angle of ~90°, smaller than that of the SNAP-II. However, its front-to-back discrimination is nearly five times better. The detector exhibits a front-to-back ratio of 33 for an AmLi source and a ratio of 9 for a ^{252}Cf source. The intrinsic detection efficiency is ~18% for a fission neutron spectrum. The absolute detection efficiency for a point source varies approximately as $1/r$ for r less than the detector dimension and as $1/r^2$ for r more than twice the detector dimension. This relationship is not valid if neutron absorbers or reflectors are present. To determine the neutron background, the detector can be rotated 180° to face away from the source of interest. Or the aperture can be covered with a thick piece of polyethylene. Four-in.-thick polyethylene reduces the neutron signal by approximately a factor of 10.

An array of slab detectors has been proposed for verification of the proper operation of centrifuge facilities (Ref. 11). Detection is possible because highly enriched uranium production is accompanied by elevated neutron levels from (α,n) reactions following ^{234}U decay in gaseous UF_6. Because of its lower mass, $^{234}UF_6$ is enriched even more than $^{235}UF_6$ in the separation process. Since the quantity of highly enriched uranium can be quite small, the increase in slab detector response can also be quite small compared with normal background. Sensitivity can be enhanced with proper design (see Chapter 14) and by using filtering and decision analysis techniques (Ref. 12).

A proposed system would consist of an array of optimized detectors within a process building having centralized data collection and processing. During normal unattended operation a minicomputer would poll the detectors remotely. Based on experimental benchmarks and Monte Carlo simulations, detection sensitivities should be adequate to detect production of highly enriched uranium. Potential sources of false alarms include normal motion of UF_6 cylinders and neutron sources used to verify criticality alarms.

15.3.2 Holdup Measurements

Slab detectors have been used for plutonium holdup measurements after a cleanout operation (Ref. 13). The model used to interpret the results assumed that the geometry of the source material could be approximated by a uniform plane near the floor. This assumption could be investigated by moving the detector and repeating the measurement. The detector was 0.23 m^2 in frontal area and unshielded. The cadmium absorbers were removed to obtain maximum sensitivity to low-energy neutrons. The advantages of using neutron techniques over gamma-ray-based techniques are that shielding of the source material is less of a problem and room return (neutrons scattered off room surfaces) tends to enhance the counter response to material at the edge of the room. These two effects reduced the number of measurement positions required to adequately survey the facility. The room-to-room shielding was good; consequently, cross talk between rooms was not a problem. The chemical and isotopic form of the plutonium was known, so that the response per gram could be determined. The detector response was averaged over several measurement positions to reduce the detector's sensitivity to hot spots (or to determine if they existed). Although this procedure yields 50% results at best, such results are adequate for applications that involve a few hundred grams of plutonium spread thinly over large areas.

15.4 THE 4π COUNTER

The 4π counter is designed to surround the sample, so that the solid angle for neutron detection approaches 4π. This configuration provides the highest possible counting efficiency and the best configuration for shielding. Shielding can be placed on all of the external surfaces of the detector to reduce the effect of nondirectional neutron backgrounds. The sample to be assayed is placed in the central well or sample chamber. Usually the 4π counter is designed to give a reasonably flat response over the volume of this chamber. This feature is very important for heterogeneous samples. Most 4π counter designs are optimized for a particular measurement application, and administrative controls are required to ensure that the samples have the appropriate fill height, composition, and matrix. If the samples are well-characterized and consistent with respect to neutron transport characteristics, assays with a few percent uncertainty are easily obtained.

The 4π counter electronics resembles a collection of slab detector electronics. Typically four to six banks of neutron detectors are used and their outputs are combined to yield the total neutron count rate. With the addition of a coincidence circuit, the same configuration can be used for coincidence counting. For either total or coincidence counting the operation of the counter is usually automated by including a calculator or computer. The computer can be programmed to control the counting electronics, convert the response to a mass measurement using a calibration function, and provide measurement control.

15.4.1 Box Counter

Figure 15.5 shows the design of a 4π counter used at the United Nuclear Corporation (UNC) in Richland, Washington, (Ref. 14) to measure end crops from the fuel extrusion process. The end crops are at least 75 wt% low-enriched uranium, the rest being zircalloy and copper. They are packaged in large shipping crates and loaded onto the counter with a fork lift. Typical net sample weights are 320 kg, however they can be as large as 550 kg. For the UNC application total neutron counting was preferred to coincidence counting for several reasons. Total counting is less sensitive to multiplication effects, such as neutron-induced fissions in ^{235}U. It is less sensitive to position-dependent variations in detection efficiency within the sample chamber. The coincidence count rate from ^{238}U is quite low, only two to five times the rate from cosmic-ray-induced events. Total counting provides a rate about 10 times the coincidence count rate, and administrative controls are readily available to ensure that only valid samples are placed in the counter.

The observed signal from the end crops is due primarily to spontaneous fission of ^{238}U. One kilogram of ^{238}U emits 13.6 n/s. The interior of the sample chamber is cadmium-lined. Under these conditions the absolute efficiency for an AmLi neutron source at the center of the chamber is 20%. The external shield is 10.2-cm-thick polyethylene. Independent measurements with a slab counter indicated that the polyethylene shielding reduces the signal from fuel stored in a neighboring room by a factor of 25. In addition, administrative controls limit the amount of uranium outside the counter: none may be placed within 8 ft of the counter and only one box may be placed within 50 ft. The counter is fully automated and the user interacts with a Hewlett

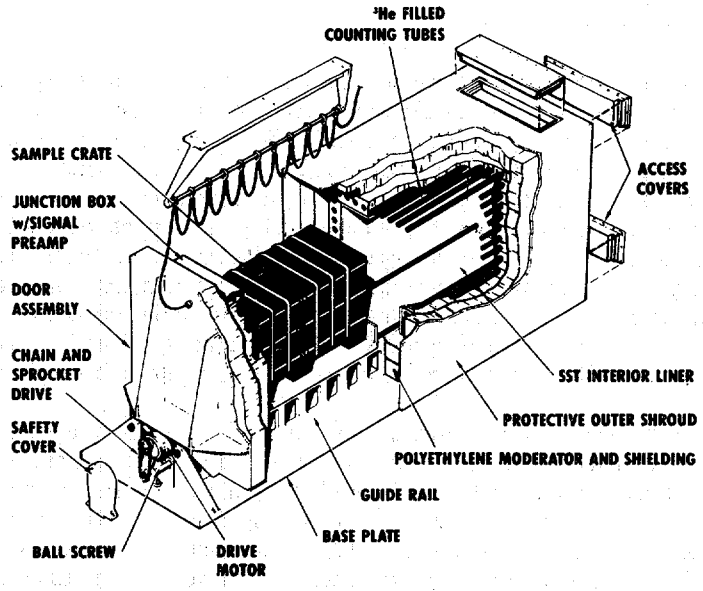

Fig. 15.5 Box counter in use at the United Nuclear Corp (UNC).

Packard HP-85 minicomputer. A daily measurement control procedure verifies that the counter is operating properly. It consists of a background count, which must be below 11 kg effective, and the assay of a standard, which must be within 2% of the known value.

Figure 15.6 is a calibration curve obtained with finished fuel from the extrusion process. Counting times of 1000 s were used; the statistical precision is better than 1%. Three types of fuel were used in the calibration: 0.947%-enriched material in 6-cm-diam cylinders (crosses); 0.947%-enriched material in 3-cm-diam cylinders, so that the packing density is nearly twice as high (circles); and 1.25%-enriched material in 6-cm-diam cylinders (x's). Two different shipping crates were used for the 1.25% material and their weight differed by 20%. The heavier crate provided a 2% lower response than the lighter crate. Assay accuracy is currently limited to 2 to 3% by variations in the shipping boxes. A limited investigation of matrix effects indicates they will be 1% or less. A preliminary comparison with end crops that were dissolved after assay indicates a bias of less than 1%.

15.4.2 Measurement of ^{238}Pu Heat Sources

A small 4π counter has been designed for the assay of ^{238}Pu heat sources at Los Alamos (Ref. 15). The heat sources are 10-g capsules of $^{238}PuO_2$. The plutonium is enriched to 83% in ^{238}Pu and the oxygen is highly enriched in ^{16}O. Heat is produced from the alpha-particle emission associated with the decay of ^{238}Pu. The strong alpha emission rate can also lead to a high neutron production rate via (α,n) reactions in ^{17}O and ^{18}O despite the depletion of ^{17}O and ^{18}O. A quantitative assay of ^{238}Pu is difficult

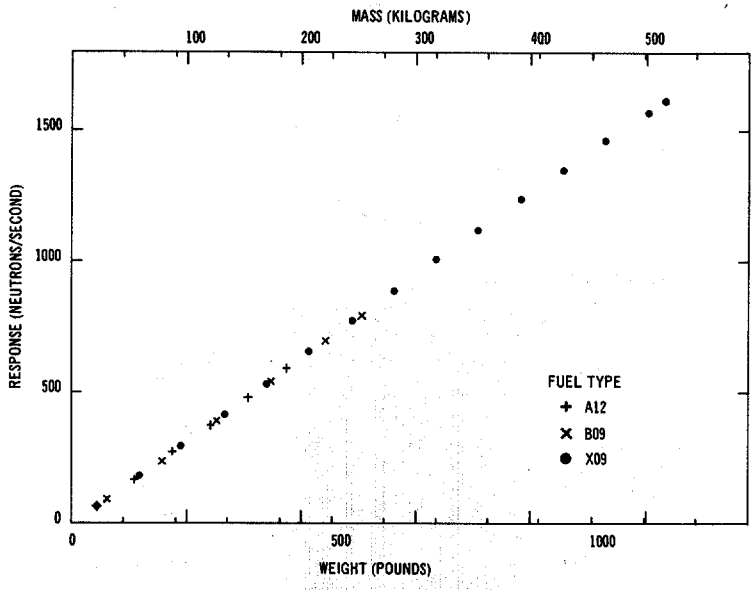

***Fig. 15.6** Calibration data for the UNC box counter showing neutron response as a function of fuel mass.*

because the exact amount of remaining ^{17}O and ^{18}O is unknown. However, the primary objective of the measurement is assurance that the neutron emission from (α,n) reactions has been reduced as low as possible. The $^{238}PuO_2$ material is handled separately from other material in the plutonium facility at Los Alamos. It undergoes different processing and exhibits the rather unique characteristic of producing heat in easily detected amounts. Consequently, administrative controls that ensure that only $^{238}PuO_2$ is being measured are easy to implement.

The heat source counter is illustrated in Figure 15.7. Its design is quite conventional, with high-density polyethylene moderator, cadmium absorber, ^3He counting tubes, aluminum outer skin, and a stainless steel and aluminum sample carrier for placing the heat source into the central counting cavity. A 10-cm-thick polyethylene shield is included to reduce the high background counting rate expected in the plutonium facility. The counter has an efficiency of 18%. Precisions of 0.5% or better are obtained in 100-s count times. The measurement accuracy is 1% within a batch of material and 5% between batches. Most of the batch-to-batch variations are due to different oxygen isotopic distributions.

15.5 MEASUREMENT OF LOW-LEVEL WASTE

Passive neutron counting is often used for measuring nuclear waste material because neutrons can penetrate large waste containers much better than gamma rays can, particularly if the waste contains dense, high-Z materials. Waste containers are typically

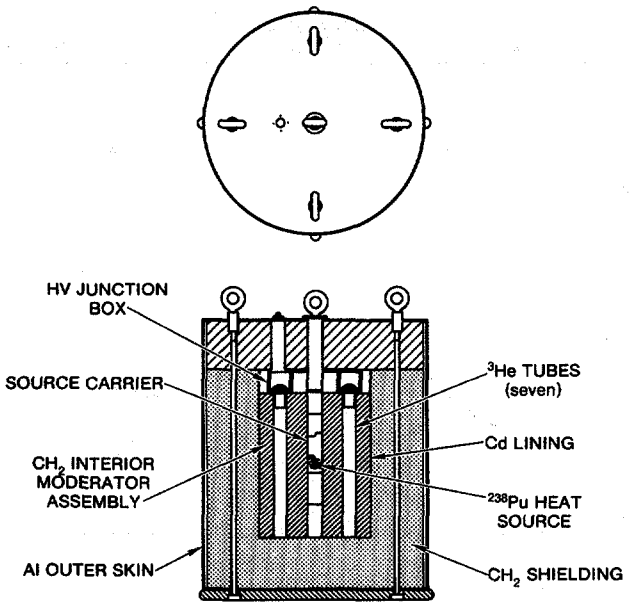

Fig. 15.7 A 4π neutron counter for ^{238}Pu heat sources (Ref. 15).

55 gallons or larger in volume, and passive gamma-ray detectors tend to underestimate the amount of nuclear material because of gamma-ray absorption in the matrix. Neutron measurements, on the other hand, tend to overestimate the amount of nuclear material; (α,n) reactions in the matrix or moderation followed by induced fission create "extra" neutrons. Neutron coincidence counting can substantially reduce this matrix sensitivity by discriminating between source fission neutrons and matrix (α,n) neutrons. However, total neutron counting may be more sensitive to small quantities of nuclear material if (α,n) reactions increase the neutron emission rate. For example, it is several orders of magnitude more sensitive to the fluorides PuF_4 or UF_6. Total neutron counting is often used for discard/save decisions rather than for quantitative assays.

15.5.1 Detection Sensitivity

The neutron emission rates of some common nuclear materials are listed in Tables 14-2, 14-3, and 14-4 (see Chapter 14). From these tables, it is clear that low-Z materials that allow (α,n) reactions can significantly increase the neutron output. Because the neutron output of the plutonium compounds is much higher than the neutron output of the uranium compounds, the sensitivity to plutonium is much better than the sensitivity to uranium. Here the sensitivity of the assay is defined as $\Delta C/C$, where ΔC is the 1-standard-deviation error in the counts C.

An important concept for the measurement of low-level waste is the detectability limit, which is that quantity of material that produces a signal that is larger than

background by the ratio $d = C/\Delta C$. For a background rate b much less than the signal rate, the detectability limit m (in grams) is given by

$$m = d^2/At_1 \quad . \tag{15-7}$$

For a background rate b much larger than the signal rate,

$$m = \frac{d}{A}\sqrt{\frac{b}{t_1} + \frac{b}{t_2}} \quad . \tag{15-8}$$

A is the response rate of the instrument in counts per second per gram, and t_1 and t_2 are the signal and background count times, respectively. Detectability limits at 3σ above background ($d = 3$) are 23 mg for low-burnup plutonium, 0.5 mg for PuF_4, 170 g for natural uranium, and 30 g for natural UF_6 (Ref. 16). These calculations assume 1000-s counting times in a large 4π counter with 15% absolute efficiency. The 4π counter is recommended for assaying low-level waste because of the weak emission rate and heterogeneous nature of the waste.

The calculated detectability limits show that passive neutron counting of low-level waste is usually practical only for plutonium. For 55-gal drums containing 100 kg of nonabsorbing matrix materials, the plutonium limit of 23 mg corresponds to about 23 nCi/g. This detectability limit can easily increase by an order of magnitude for actual drums that contain significant quantities of moderators or neutron poisons. In most cases, however, passive neutron assay overestimates the quantity of nuclear material present because of (α,n) reactions in the matrix. Unless the chemical and isotopic form of the waste is known, no quantitative conclusion can be drawn about the nuclear content of a barrel other than an upper limit.

15.5.2 Assay of 55-Gallon Drums

Total neutron counting of 55-gal drums containing PuO_2-contaminated waste has been investigated at Los Alamos (Ref. 17). Measured were a set of 17 standards constructed at the Rocky Flats Plutonium Processing Facility (Ref. 18). The standards were designed to simulate the contaminated process materials and residues routinely assayed in the Rocky Flats drum counter. Table 15-2 summarizes the characteristics of these drums. A standard deviation of $\pm 16\%$ was obtained for the 17 drums. Since the plutonium isotopics and chemical form were both fixed and well known, the total neutron signal was a reasonable measure of the plutonium content.

15.5.3 Assay of Large Crates

A neutron counter large enough to assay 1.2- by 1.2- by 2.1-m waste crates was developed and used extensively at the Rocky Flats facility (Ref. 19). This 4π counter uses twelve 30-cm-diam ZnS scintillators spaced around the sample chamber. Because these fast-neutron detectors also exhibit some gamma-ray sensitivity, the discriminator thresholds are set above the 1.3-MeV ^{60}Co gamma-ray energy. Most of the neutron

Table 15-2. Physical characteristics of Rocky Flats 55-gallon drum standards

Description	Matrix	Matrix Composition (wt%)	Matrix Av Net Weight (kg)	Matrix Av Density (g/cm^3)	Plutonium (as PuO_2) Loadings (g Pu)
Graphite molds	60-mesh graphite	100	110	0.53	60, 145, 195
Dry combustibles	carbon	90	24	0.12	10, 165, 175
	plastics	5			
	cellulose	5			
Wet combustibles	cellulose	80	51	0.25	28.5, 166
	water	15			
	plastics	5			
Washables	polyvinyl	42	32	0.15	10, 90, 160
	lead gloves	28			
	polyethylene	20			
	cellulose	7			
	surgical gloves	3			
Raschig rings	Pyrex glass with 12% boron as B_2O_3	100	82	0.39	40, 95, 185
Resin	Dowex 1 × 4	100	–	–	25, 110
Benelex-Plexiglas	–	–	–	–	75

signal is also discriminated out, and the measured efficiency of the counter is 0.1%. Although the desired signal is from spontaneous fission neutrons, total counting is preferred to coincidence counting because of the low detection efficiency.

The major sources of inaccuracy for the crate counter are variable matrix effects and the unknown chemical form of the plutonium. Some comparisons with a 55-gal-drum counter show that the crate counter tends to overestimate the plutonium content. Typical crate loadings are less than 100 g; the counting times are 200 s. The results typically agree with tag values within a factor of 4.

The crate counter is used to flag crates that need to be opened and checked more carefully. It periodically locates crates that have been labeled incorrectly. The counter is also used in conjunction with passive gamma-ray counting if more quantitative results are desired.

15.6 SPECIAL APPLICATIONS

The special applications described in this section focus on the neutron energy spectrum observed by some passive assay instruments. In the first two examples, the observed energy spectrum is exploited to determine a specific feature of the sample.

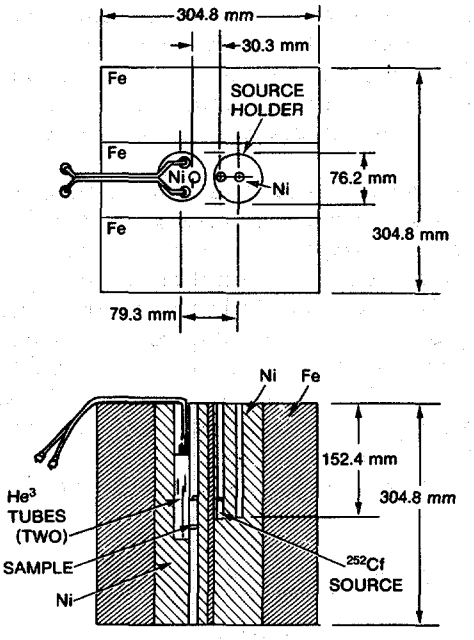

Fig. 15.8 A ^{252}Cf-based hydrogen analyzer.

15.6.1 The ^{252}Cf Hydrogen Analyzer

The ^{252}Cf hydrogen analyzer (Figure 15.8) can determine the hydrogen content of small uranium samples by measuring the softening of the neutron spectrum of a nearby ^{252}Cf source (Ref. 20). The analyzer consists of a steel block with holes bored for two 76-mm-diam nickel cylinders. One cylinder contains the ^{252}Cf source, and the other holds the sample and two ^{3}He neutron detectors, as detailed in Figure 15.9. Because ^{3}He tubes have a higher efficiency for low-energy neutrons, the count rate increases for samples containing hydrogen. This matrix effect is enhanced by using steel reflectors instead of polyethylene moderators around the sample and detectors.

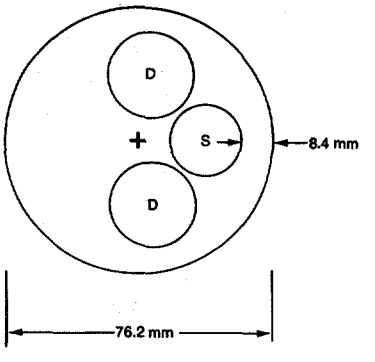

Fig. 15.9 Expanded view of the sample-detector geometry in the ^{252}Cf hydrogen analyzer. The detectors (D) are 25 mm in diameter and the sample chamber (S) is 19 mm in diameter.

The counting electronics requires only a single-channel analyzer. A californium source strong enough to give a count rate of 10 kHz is used so that 0.1% precision can be obtained in reasonable counting times (100 s). Long-term drifts of 0.3% were observed over a 3-day period; however, the actual data consists of a sample measurement preceded or followed by a background measurement. Consequently, long-term drifts are not a problem.

Figure 15.10 demonstrates the response (difference between sample and background counts) as a function of increasing hydrogen. The straight line is a least squares fit to the data. The four samples indicated in the graph had a graphite matrix and were of equal volumes. Figure 15.11 shows the effect of increasing ^{235}U content on the hydrogen analyzer. The hydrogen analyzer was originally designed for use in conjunction with a Small Sample Assay System (SSAS)(Ref. 21) to determine the uranium content of moist uranium fuel rods. The SSAS determines uranium content, but the result is affected by hydrogen content. Consequently an iterative procedure is employed to obtain the best possible assay. First, the SSAS measurement provides a value for uranium content good to about ±10%. Then the hydrogen analyzer results can be used to determine the hydrogen content to ±2 mg, and finally the SSAS result can be iterated to determine ^{235}U to better than 1% accuracy.

15.6.2 Moisture Determination by Detector Ring Ratio

The ratio of total neutron counts in the inner and outer detector rings of a two-ring 4π counter is a measure of the neutron energy spectrum. In a way similar to the ^{252}Cf hydrogen analyzer, the ring ratio can provide information about the moisture content of the sample.

This moisture monitoring technique has been applied to the assay of wet plutonium oxalate (Refs. 22 and 23). The oxalate is precipitated during the conversion of plutonium nitrate to oxide. It is heterogeneous and typically contains 30 to 65 wt% water. The

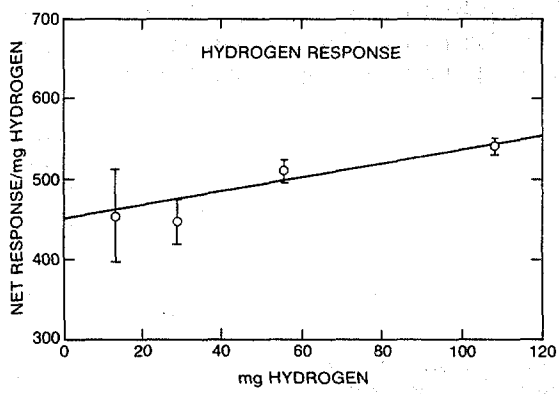

Fig. 15.10 *Net signal per milligram of hydrogen as a function of hydrogen content.*

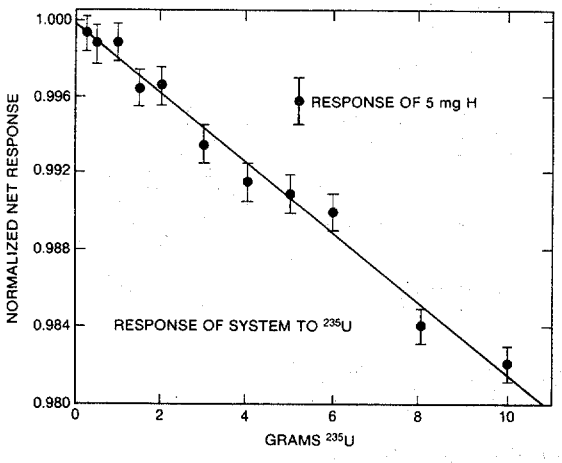

Fig. 15.11 *Effect of ^{235}U on the hydrogen analyzer.*

plutonium assay is done by passive neutron coincidence counting, but a correction for water content is based on the ring ratio. The correction is needed because hydrogen is a better moderator than plutonium oxalate and samples with higher water content decrease the average neutron energy more. The neutron counter contains an inner ring of detectors separated from the sample by 11 mm of polyethylene and an outer ring separated by 33 mm of polyethylene. A change in the neutron energy spectrum affects the two rings differently because of their different depth in the polyethylene.

The counter was calibrated by assaying 19 samples of known plutonium and water content. The mass m of plutonium was expressed as

$$m = aR_0^{(\alpha+\beta T_i/T_o)} \tag{15-9}$$

where R_0 = coincidence count rate in the outer ring
T_i = total count rate in the inner ring
T_o = total count rate in the outer ring
and a, α, β are fitted parameters.

An additional 22 samples were assayed nondestructively with a standard deviation of 2.2% (1σ)(Ref. 22) relative to later destructive analyses. Without the ring ratio correction for moisture, the deviation is 50 to 100%.

15.6.3 Energy-Independent Long Counter

There are many uses in fast-neutron studies for a counter whose detection efficiency is independent of the incident neutron energy. One such counter used to calibrate and standardize neutron assay instruments is the long counter (Refs. 10 and 24). The energy response of the long counter is nearly flat, but not entirely energy-independent, as has sometimes been assumed.

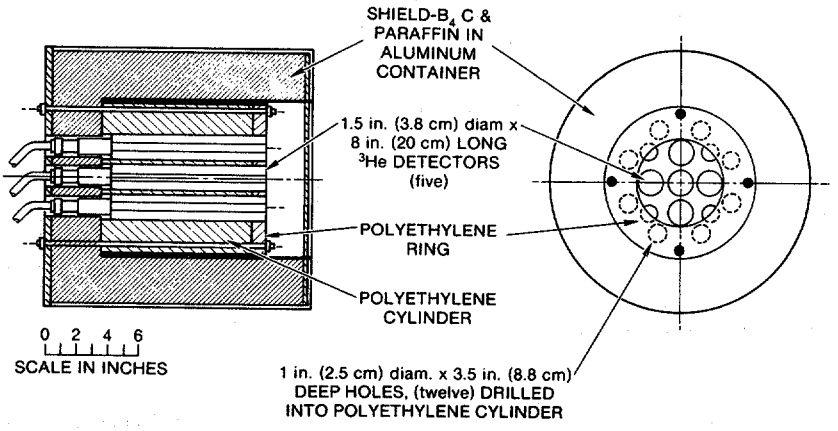

Fig. 15.12 A high-efficiency ^{3}He long counter.

Figure 15.12 shows the design features of a high-efficiency long counter. The neutron source is aligned on the axis of the center ^{3}He detector, at a distance of at least 1 m. The minimum source-to-detector distance, the number and location of the holes in the moderator, and the design of the polyethylene ring in front of the moderator are all essential for achieving the flattest energy response. Figure 15.13 (Ref. 10) shows the response of the counter to broad-spectrum neutron sources; the relative source strengths were known to 3%. Within the accuracy of the source strengths, the counter response appears to be flat from 0.024 MeV to more than 4 MeV. More recent data (Figure 15.14) were obtained with the same counter using a Van de Graaff accelerator to provide relatively monoenergetic neutrons from the ^{7}Li(p,n)^{7}Be reaction (Ref. 25). As the neutron energy was varied from 100 to 1200 keV, a small resonance was observed at 450 keV. Clearly the counter response is not completely energy-independent, but only approximately so.

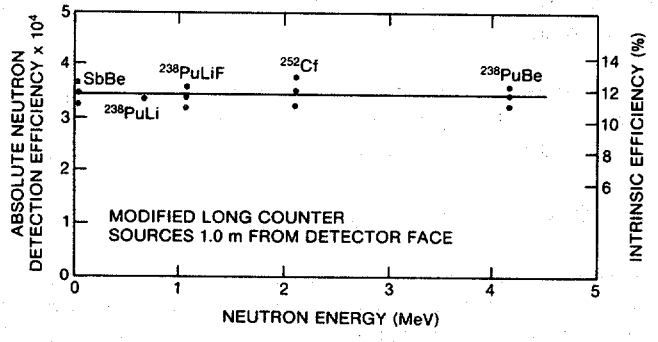

Fig. 15.13 Efficiency of the ^{3}He long counter as a function of neutron energy, as determined with radioactive sources.

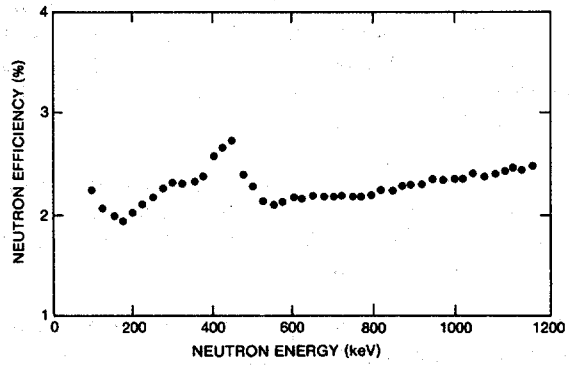

Fig. 15.14 *Efficiency of the ^{3}He long counter as a function of neutron energy, as determined with mono-energetic neutrons.*

REFERENCES

1. H. O. Menlove, T. L. Atwell, and A. Ramalho, "Upgrading of SNAP Neutron Detector, SNAP-II," in "Nuclear Safeguards Research Program Status Report, September—December 1975," Los Alamos Scientific Laboratory report LA-6316-PR (April 1976), pp. 5-6.

2. R. B. Walton and T. L. Atwell, "Portable Neutron Probe, 'SNAP'," in "Nuclear Analysis Research and Development Program Status Report, January—April 1973," Los Alamos Scientific Laboratory report LA-5291-PR (May 1973), p. 14.

3. T. L. Atwell, D. B. Smith, and A. C. Walker "Assay of Plutonium Metal Buttons with a Portable Neutron Counter," in "Nuclear Analysis Research and Development Program Status Report, May—August 1973," Los Alamos Scientific Laboratory report LA-5431-PR (November 1973), pp. 20-23.

4. R. B. Walton, T. D. Reilly, J. L. Parker, J. H. Menzel, E. D. Marshall, L. W. Fields, "Measurement of UF_6 Cylinders With Portable Instruments," *Nuclear Technology* 21, 133 (1974).

5. NRC Guide 5.23, "In-Situ Assay of Plutonium Residual Holdup," Rev. 1 (February 1984).

6. C. Kindle, "In-Situ Measurement of Residual Pu," *Nuclear Materials Management* V, 540 (1976).

7. R. B. Walton, "The Feasibility of Nondestructive Assay Measurements in Uranium Enrichment Plants," Los Alamos Scientific Laboratory report LA-7212-MS (April 1978).

8. J. T. Caldwell, S. W. France, R. D. Hastings, J. C. Pratt, E. R. Shunk, T. H. Kuckertz, and R. Goin, "Neutron Monitoring of Plutonium Storage Vaults," in "Nuclear Safeguards Research and Development Program Status Report, October 1980—January 1981," C. N. Henry, Comp., Los Alamos National Laboratory report LA-8870-PR (November 1981), pp. 21-28.

9. E. J. Dowdy, A. A. Robba, and R. D. Hastings, "Power Reactor Monitor," in "Nuclear Safeguards Research and Development Program Status Report, May—August 1978," S. D. Gardner, Ed., Los Alamos Scientific Laboratory report LA-7616-PR (April 1979), pp. 58-60.

10. L. V. East and R. B. Walton, "Polyethylene Moderated ^{3}He Neutron Detectors," *Nuclear Instruments and Methods* 72, 161 (1969).

11. J. E. Stewart, S. C. Bourret, R. W. Slice, J. T. Markin, C. A. Coulter, and C. A. Spirio, "An Area Neutron-Monitoring Strategy for International Inspections at Centrifuge Enrichment Plants," in "Safeguards and Security Status Report, February—July 1982," Los Alamos National Laboratory report LA-9595-PR (February 1982), pp. 53-56.

12. J. T. Markin, J. E. Stewart, and A. S. Goldman, "Data Analysis for Neutron Monitoring in an Enrichment Facility," European Safeguards Research and Development Association Specialists' Meeting, Petten, The Netherlands, April 27-29, 1982, Los Alamos National Laboratory document LA-UR-81-2304.

13. J. W. Tape, D. A. Close, and R. B. Walton, "Total Room Holdup of Plutonium Measured With a Large-Area Neutron Detector," *Nuclear Materials Management* V (3), 533 (1976).

14. J. K. Sprinkle, Jr., and M. M. Stephens, "End-Crop Box Counter Manual," Los Alamos National Laboratory report LA-9781-M (June 1983).

15. C. O. Shonrock, L. G. Speir, P. R. Collinsworth, and N. Ensslin, "Neutron Counter for ^{238}Pu Heat Source Program," in "Nuclear Safeguards Research and Development Program Status Report, April—June 1980," Los Alamos Scientific Laboratory report LA-8514-PR (February 1981), pp. 5-6.

16. T. Crane, "Measurement of Uranium and Plutonium in Solid Waste by Passive Photon or Neutron Counting and Isotopic Neutron Source Interrogation," Los Alamos Scientific Laboratory report LA-8294-MS (March 1980).

17. T. D. Reilly and M. M. Thorpe, "Neutron Assay of 55-Gallon Barrels," in "Nuclear Safeguards Research and Development Program Status Report, May—August 1970," Los Alamos Scientific Laboratory report LA-4523-MS (September 1970), pp. 26-29.

18. L. Doher, "A Measurement Control Scheme for Plutonium Counting Systems," Proc. Institute of Nuclear Materials Management Annual Meeting, Gatlinburg, Tennessee (May 1970).

19. R. Harlan, "U and Pu Assay of Crated Waste by Gamma-ray, Singles Neutron, and Slow Neutron Coincidence Counting," *Measurement Technology for Safeguards and Materials Control*, T. R. Canada and B. S. Carpenter, Eds. (NBS Special Publication 582, 1980).

20. D. A. Close, R. C. Bearse, and H. O. Menlove, "^{252}CF-Based Hydrogen Analyzer," *Nuclear Instruments and Methods* 136, 131 (1976).

21. A. E. Evans, Jr., *IEEE Transactions on Nuclear Science* 21 (1), 628 (1974).

22. T. W. Crane, "Calibration of a Neutron Coincidence Counter for Measurement of the Plutonium Content of Wet Oxalate Cakes," Los Alamos National Laboratory report LA-9744-MS (April 1983).

23. R. S. Marshall and T. R. Canada, "An NDA Technique for the Assay of Wet Plutonium Oxalate," *Nuclear Materials Management* 107-113 (1980).

24. W. D. Allen, *Fast Neutron Physics*, J.B. Marson and J. L. Fowler, Eds. (Interscience Publications, 1960), Vol. I, p. 361.

25. A. E. Evans, Jr., "Energy Dependance of the Response of a ^{3}He Long Counter," *Nuclear Instruments and Methods* 199, 643 (1982).